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Development of Delayed Gamma-ray Spectroscopy for Nuclear Safeguards (2): Designing a Compact DGS Instrument

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ABSTRACT

The Japan Atomic Energy Agency is developing the Delayed Gamma-ray Spectroscopy (DGS) non-destructive assay technique to quantify the fissile-nuclide content in small samples of mixed nuclear materials. One of our primary goals is to develop a compact and efficient DGS instrument to be easily installable into analytical laboratories. The instrument should include an external neutron source and a gamma-ray detection system along with other supporting systems like sample transfer and neutron monitoring. One of the challenges is to design a compact and efficient moderator for commercial neutron sources (e.g. neutron generators or sealed radioactive sources) that emit neutrons with high energy. However, to be able to enhance the gamma-ray signal from fissile materials, thermal neutrons are best due to their higher fission cross-sections. The choice of viable neutron source (neutron spectrum and strength) depends on several considerations (e.g. sample type and interrogation pattern), but also affect the gamma-ray measurement and the consequence analysis. In this work, we will first describe the evaluation results of our Delayed Gamma-ray Test Spectrometer using a ^{252}Cf source (DGTS-C) from the first experiment carried out in PERLA in collaboration with the European Commission, Joint Research Centre. In association, we will describe how it provided guidance for our demonstration irradiator. Further, we will present our final moderator design concept for a deuterium-deuterium (D-D) neutron generator and present the latest results of data-model comparisons, including those with our PUNITA results.

INTRODUCTION

For non-nuclear weapon states, safeguards agreements [1] are required under the Non-Proliferation Treaty [2]. Special fissionable materials (e.g. ^{239}Pu, ^{233}U, ^{235}U) and source materials (i.e. depleted and natural U, Th) need to be verified to prevent diversion. All of these are commonly found in spent fuel, but mostly within full assemblies that are sealed and verified as an item. However, spent nuclear fuel in reprocessing plants is converted into a solution form, requiring the material to be verified through sampling. These samples are verified using Hybrid K-Edge Densitometry (HKED) active-interrogation non-destructive assay (NDA) to quantify the elemental U and Pu content [3]. The nuclide content of some samples is determined by Isotope Dilution Mass Spectrometry (IDMS) [4]. This destructive analysis (DA) technique reaches high precision, but requires long sample preparation that extends the reporting time [5]. Moreover, it produces waste in the laboratory and consumes reference materials that are becoming limited in availability.

The Integrated Support Center for Nuclear Nonproliferation and Nuclear Security (ISCN) of the Japan Atomic Energy Agency (JAEA) is investigating methods to improve spent nuclear fuel solution verification. In particular, JAEA/ISCN is developing the Delayed Gamma-Ray Spectroscopy (DGS) active NDA technique with the intent to verify the fissile nuclide composition. For small samples of spent nuclear fuel solutions, JAEA/ISCN is working on the design and validation of a compact instrument to be easily installable in analytical laboratories. A DGS instrument consists of an irradiator to perform neutron-induced fission and a detection system to observe the gamma rays emitted by the generated fission products. Due to the intense radiation emitted by long-lived fission products (^{137}Cs) in spent nuclear fuel, a detector filter is required to reduce the count rate. After irradiation, the gamma rays above 2.5 MeV emitted by the short-lived fission products can be collected in a gamma detector. To compensate for the signal reduction caused by the filter, the source intensity must be increased. Commercially available neutron sources emit high energy neutrons that require moderation. In this way, the fissile fission rate is increased due to the high fission cross-section in the thermal energy range.

Starting in 2015, the JAEA/ISCN performed several fundamental studies in collaboration with the European Commission, Joint Research Centre (EC/JRC) using PUNITA [6, 7] in the Ispra (Italy) site. Originally designed for differential die-away studies, PUNITA was modified to be able to conduct DGS experiments [8]. Combining lessons learned from these experiments and MCNP [9] models, the JAEA/ISCN designed the Delayed Gamma-ray Test Spectrometer (DGTS) with the usage of a ^{252}Cf source (DGTS-C). Starting in February, 2018, the DGTS-C was tested in PERLA of JRC-Ispra. In this paper we describe the current design of the DGTS-C showing how irradiator and detector work together in a compact instrument. We will then present the latest results in comparison with the ones collected using PUNITA. Finally, we will describe how all the results can be used and combined together to design a compact and practical instrument. Finally, we will describe how all the results can be used and combined together to design a compact and practical instrument.

DGTS-C

The main goal of the JAEA/ISCN instrument development was to show the practicality of having a compact instrument for DGS application. Based on a preliminary ^{252}Cf moderator model study [10], it was initially designed with the sample being in a fixed position (static configuration of the DGTS-C) due to facility constraints. However, due to changes in the test location the instrument was tested in PERLA and adapted to test the available U and Pu certified standard samples [11] with maximum dimensions of 10x10x15 cm^3. Moreover, the usage of these reference materials allows for a transient configuration of the instrument with the sample moving between irradiation and measurement positions (see Figure 1). The goal was to show the feasibility of a compact DGS instrument.

Intended to be used with ^{252}Cf sources, the irradiator was designed with overall dimensions of 75x55x70 cm^3. The choice of using ^{252}Cf was firstly driven by the lower energy neutrons compared to those emitted by a deuterium-tritium (D-T) generator (as used in PUNITA). Additionally, the ^{252}Cf source was more readily available than a deuterium-deuterium (D-D) generator that emits neutrons of similar energy. Even with the lower starting energy, the emitted neutrons require moderation to reach the thermal energy regime and reduce the dose rate. This was performed by inserting the source into a high-density polyethylene (HDPE) thermalizer, using graphite to retain and reflect the neutrons toward the sample, and a final HDPE shell outside the graphite.

The detector used in our tests was a liquid nitrogen cooled, 50% efficiency high-purity germanium

(HPGe). The detector was placed far from the Cf source to minimize neutron damage with a thick shield consisting of Pb, HDPE and Cd layers around the detector head. The distance between the sample and the detector was kept short with a thin Pb filter used to reduce the low energy gamma rays coming from the sample. To obtain the spectra, a LYNX digital signal analyzer [12] was used in the multispectral scaling (MSS) mode.

For the uranium samples, we used four different ^{235}U enrichments samples from the Central Bureau for Nuclear Measurements (CBNM) from natural uranium to about 5.0% enrichment. As a reference sample, we used a dummy aluminum CBNM. For the Pu samples, we combined five different PuGa alloy disk samples. These were the same disks used in PUNITA, but were spread in a 2x2 matrix with the fifth one slightly overlapping in the center rather than in a stack. In the shuttle the sample was surrounded by HDPE, with a graphite infill to improve neutron moderation and retention. An HDPE tunnel shield with Cd liners was installed to reduce the background from the instrument activation and to further reduce dose rates.

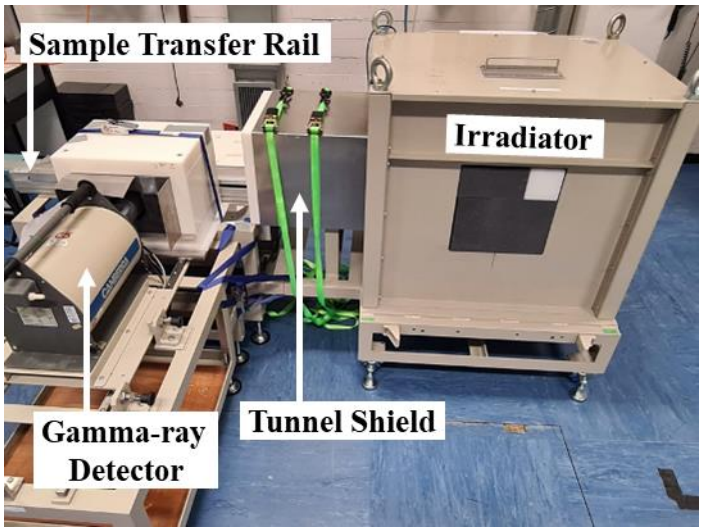

Figure 1: Transient configuration of the JAEA/ISCN DGTS-C.

CURRENT EXPERIMENT RESULTS

The irradiator was designed to use a ^{252}Cf source that emits neutrons with mean energy around 2 MeV. Compared to the D-T neutron generator used in PUNITA, the lower emission energy doesn't require heavy materials (i.e. lead, tungsten) for first-stage moderation. With this and the smaller size of the ^{252}Cf source, the distance between the source and the sample was able to be reduced. Neutron measurements in the DGTS-C using a small ^{3}He detector confirmed how increasing the distance with the source results in a decreasing number of counts in the detector [13]. Moving the ^{3}He detector vertically showed that we could allign it with the source, following decrease in the number of counts, allowing us to optimize the greatest thermal flux. Additional short-distance studies were performed in collaboration with the EC/JRC, evaluating the flux obtained in a full HDPE moderator with activation foils and validated by MCNP models [14]. Moreover, due to the lower neutron energy, the 2753 keV peak from the $^{27}Al(n,\alpha)^{24}Na$ reaction interfering with the important delayed gamma rays observed in the PUNITA spectra was not visible.

While the lower energy is beneficial, the continuous emission of the ^{252}Cf source considerably increased the instrument activation gamma-ray background. Further, the continuous emission from the source increases the dose during manipulation for the operator. Adding thicker shielding around the detector and keeping it far from the irradiator significantly reduced this background.

Moreover, adding distance improved the life expectancy of the detector by reducing the fast neutron damage possibility [15]. Compared to the initial static configuration, this transient version allowed the source to stay inside the irradiator to reduce the dose for the operator. The tunnel shield that helped reduce the dose during source manipulation had the additional benefit of attenuating the fast neutrons before reaching the detector.

To enhance the delayed gamma-ray signal above the remaining activation background, an MSS acquisition was implemented. MSS provides instantaneous dead-time for each 1-s integrated spectrum during the measurement periods; a benefit over the pulse-height acquisition used in PUNITA experiments. For example, during the 300-s measurement periods, the dead-time was ~30% at the beginning and ~15% at the end of each period (see Figure 2). This allows for an improved dead-time correction to better enhance the signature from the shortest-lived fission products. The dead-time corrected spectra also included the presence of several escape peaks, observed due to the 50% efficiency of the detector. This delayed gamma-ray signature, including the escape peaks, is, therefore, affected by the detector choice; we observed fewer escape peaks with the 80% efficiency detector in PUNITA spectra.

Regardless, after applying this dead-time correction, gamma-ray peaks were observed with intensities similar to PUNITA, even with an ~80% lower neutron source intensity. Notably, an excess in the Pu primary peak intensities were observed in the DGTS-C due to the different configuration of the same samples: PUNITA used a stack; DGTS-C used a spread formation. This further confirms that sample shape needs to be considered in the design of the instrument.

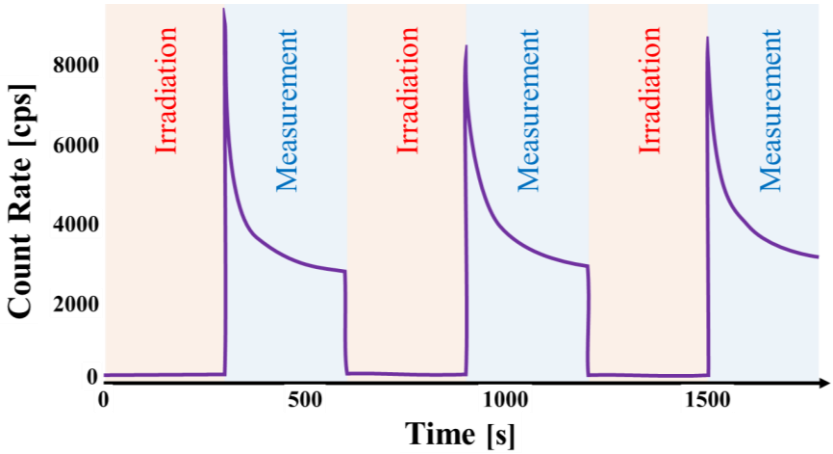

Figure 2: Representation of the LYNX MSS acquisition during a DGS measurement.

FUTURE

Applying lessons learned from both PUNITA and DGTS-C experiments, the JAEA/ISCN designed and fabricated an improved DGTS irradiator, including PERLA operational conditions. Preliminary evaluations of the new irradiator show an improved thermal neutron flux in the sample area using ^{3}He detectors and MCNP models. This was possible due to reducing the distance between sample and source; reducing the first-stage HDPE moderation; and adding thick HDPE walls on all sides, while still maintaining compactness. The thick HDPE walls further reduced the dose rate, as did the addition of better source handling using an automatic crane, as requested by the JRC. Finally, positions for ^{3}He detectors were introduced to evaluate neutron monitoring and source scaling for mass determination. This new irradiator will be used for the demonstration of DGS at the end of the current project phase (February/March 2022) and is called Delayed Gamma-ray Demonstration Spectrometer (DGDS).

PUNITA, DGTS-C, and DGDS all show gradual improvements on the general DGS instrument design requirements. The design of the final DGS instrument for small samples of spent nuclear fuel solution will incorporate both sample and facility constraints. To overcome the required gamma-ray filter, the neutron source intensity must be at least 10^9 n/s [16], indicating that the equivalent 100 GBq ^{252}Cf is not optimal due to the high dose and background [10]. A preliminary JAEA/ISCN MCNP optimization study shows that D-D neutron generators can be a viable alternative [16]. Since D-D produces neutrons with energy similar to ^{252}Cf, similar moderation efficiency is obtained. Moreover, as a pulsed source similar to D-T generators, no activation background would be produced during the measurement and low dose rates are achievable at all times. To validate the MCNP models, the neutron flux of a test moderator will be studied at the MONNET tandem beam line of the EC/JRC in Geel (Belgium). Using ^{3}He detectors, various configurations of graphite and HDPE will be tested for changing trends in the flux. Present studies are underway at the JAEA/ISCN using activation foils and ^{252}Cf for a direct comparison between the sources to establish the best DGS condition.

In combination with the external neutron source, the other critical component of the irradiator in a practical DGS instrument is the sample size and geometry. Since the current target is to apply DGS to spent nuclear fuel solution samples, discussions are underway with the EC/JRC to study properly mixed nuclear materials. Additionally, since facility constraints may require a fixed sample to reduce the contamination risk, further studies will be performed regarding how to improve early DGTS results.

SUMMARY

The JAEA/ISCN is developing a DGS instrument to quantify the fissile-nuclide content in small samples of mixed nuclear materials. Several experiments were performed in collaboration with the EC/JRC using PUNITA and the JAEA/ISCN compact DGTS-C instrument. Tested in PERLA, the DGTS-C successfully proves that a compact instrument can provide a DGS signature from nuclear material samples. Fission product gamma-ray spectra from standard reference materials produced peaks with similar counts to those obtained with PUNITA, even with a less intense neutron source. From lessons learned, the JAEA/ISCN designed and fabricated an irradiator with improved flux and source handling to reduce the dose, along with an introduction of neutron monitoring. This new irradiator is being characterized in the JAEA/ISCN laboratories and will be used to demonstrate the DGS technique during the JAEA/ISCN workshop planned for early 2022. Concurrently, the JAEA/ISCN and the EC/JRC are planning experiments at the MONNET beam line to validate MCNP models for a D-D neutron source. Finally, in the next phase project, a final practical DGS instrument will be designed and tested for the ability to quantify the fissile nuclide content in small mixed nuclear samples.

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